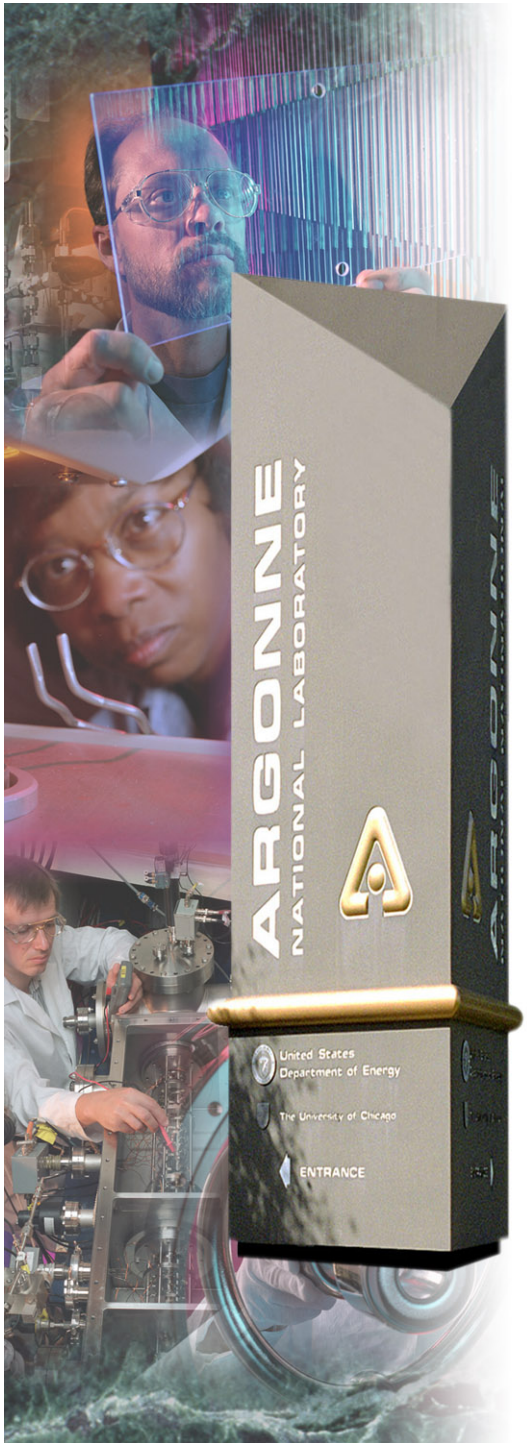

Integral Nuclear Data Analysis for Cross Section Validation

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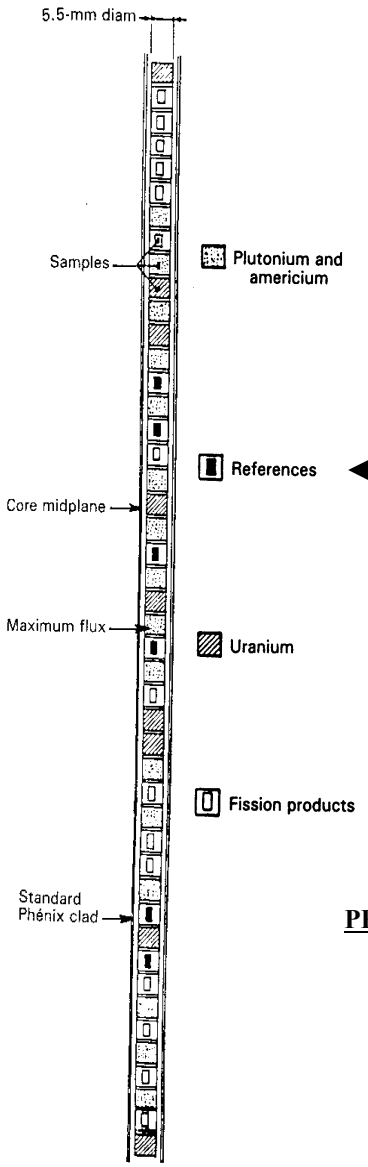
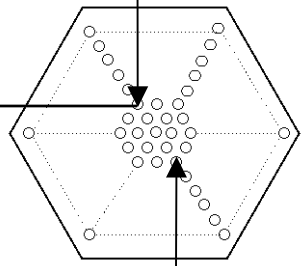
- The fuel form to be used in advanced nuclear systems dedicated to transmutation will contain a high fraction of Minor Actinides (MA). Good quality cross section data are therefore required for these isotopes in order to provide a reliable neutronic design. Basic data are available for these isotopes but still a validation is needed in order to quantify their reliability. This is traditionally done through the use of mock up experiments. The information that can be gathered on MA from experiments come mostly from small sample irradiation, reactivity oscillation, and fission and capture rates measurements
- In the framework of the CEA/DOE international collaboration on the AFC program, clean and very useful irradiation experiments have been made available:
  - **PROFIL: small sample irradiation experiment**
  - **TRAPU: fuel pin irradiation experiment**

- The first part of the PROFIL irradiation campaign, was performed at the PHENIX fast reactor in 1974 during the first three cycles
- A pin containing 46 separated capsules of pure isotopes (U, Pu, Am and fission products) was located in a standard subassembly in the first row of the inner core of PHENIX (→ clean irradiation conditions) and irradiated for 189.2 days
- A second experiment, PROFIL-2, was performed during the 18^{th}, 19th, and 20^{th} cycle of PHENIX. It contained 2x42 capsules including Neptunium samples.
- The TRAPU experiment consisted of a six-cycle irradiation of mixed-oxide pins that contained plutonium of different isotopic compositions but heavily charged in the higher isotopes (Pu-240, Pu-241 and Pu-242). Three types of plutonium containing pins were used.



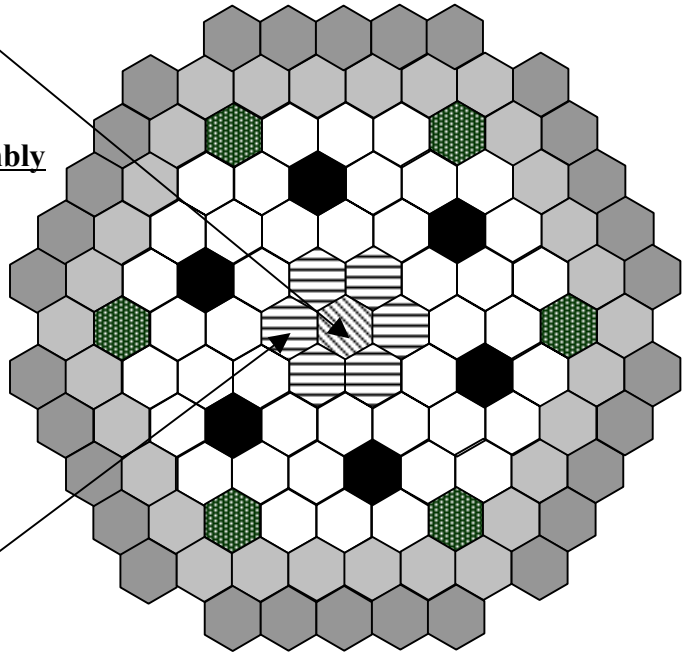
PROFIL-1 Irradiation Pin

PHENIX Subassembly



PROFIL-2 Irradiation Pin

PHENIX Core



Core 1

Core 2

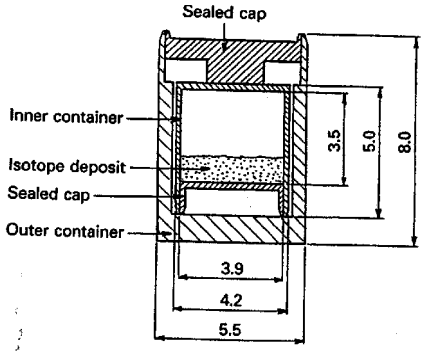
Core 3

Control Rod

Core 1

Core 1

PROFIL-1 Irradiation Pin



- **The information that can be gathered from the PROFIL post-irradiation analysis is related to the evaluation of reaction rates (capture and n,2n) for a given isotope**

$$R_c(A) = \sigma_c(A).\tau \cong \frac{N_{A+1}(\tau)}{N_A(\tau)} - \frac{N_{A+1}(0)}{N_A(0)}$$

$$R_{n,2n}(A) = \sigma_{n,2n}(A).\tau \cong \frac{N_{A-1}(\tau)}{N_A(\tau)} - \frac{N_{A-1}(0)}{N_A(0)}$$

- **The experimentally measured concentrations of neodymium were used to correctly normalize the depletion calculations and reduce the uncertainty in the irradiation history.**
- **Post-irradiation examinations were performed with spectroscopic techniques, with simple or double isotopic dilution and well-characterized tracers to measure concentrations (maximum experimental uncertainty in the order of 2 ÷ 3 %).**

- A sensitivity and uncertainty analysis relative to the impact of basic data (microscopic cross section and decay constants) on the calculated isotope concentrations involved in the evaluation of the C/E's has been performed, to understand if discrepancies in the C/E's are consistent with current uncertainties associated to nuclear data

- The GPT (Generalized Perturbation Theory) formulation has been used:

$$\frac{d\bar{n}(t)}{dt} = [A]\,\bar{n}(t)$$

$$-\frac{d\bar{n}^{*}}{dt} = [A]^{T}\bar{n}^{*}$$

;

$$S_j^K = \frac{\partial n_F^K}{\partial \sigma_j}\cdot\frac{\sigma_j}{n_F^K} = \frac{1}{n_F^K}\int_{t_0}^{t_f} \bar{n}^{*}\,\sigma_j\,\bar{n}\,dt$$

;

$$I_N = \left(\sum_{ji} S_j^n\,\overline{\overline{C_{ij}}}\,S_i^n\right)^{1/2}$$

Cross Section Uncertainty Values (%)

Isotope	ν	σ_f	σ_{inel}	σ_{el}	σ_{capt}	$\sigma_{n,2n}$
Pu-239	0.75	3.7	10	5	8.5	25
Pu-240	1.2	5	15	10	30	16
Pu-241	0.9	5	15	10	20	20
Pu-242	1.2	5	15	10	30	25
U-235	0.65	2	5	8	12	9
U-238	1.5	2.25	9.5	5	10	9
Am	5	20	50	5	40	25
Cm						
Np-237						

Data Type	C/E						A-priori Uncertainty
	JEF2.2		ENDF/B-V		ENDF/B-VI		
	PROFIL-1	PROFIL-2	PROFIL-1	PROFIL-2	PROFIL-1	PROFIL-2	
σ_{cap} U-235	0.95 ± 1.7 %	0.97 ± 1.7 %	1.00 ± 1.7 %	1.01 ± 1.7 %	0.95 ± 1.7 %	0.98 ± 1.7 %	**3.4 %**
σ_{cap} U-238	0.99 ± 2.3 %	1.01 ± 2.3 %	1.03 ± 2.3 %	1.04 ± 2.3 %	1.00 ± 2.3 %	1.02 ± 2.3%	**1.7 %**
σ_{cap} Pu-238	0.97 ± 4.0 %	1.05 ± 4.0 %	1.41 ± 4.0 %	1.45 ± 4.0 %	1.91 ± 4.0 %	1.97 ± 4.0 %	**23.6 %**
σ_{cap} Pu-239	0.97 ± 3.0 %	0.96 ± 3.0 %	0.95 ± 3.0 %	0.98 ± 3.0 %	0.93 ± 3.0 %	0.96 ± 3.0 %	**5.8 %**
$\sigma_{n,2n}$ Pu-239	0.63 ± 15.0 %	0.58 ± 15 %	0.56 ± 15.0 %	0.53 ± 5.0 %	1.05 ± 15 %	0.91 ± 15.0 %	**11.7 %**
σ_{cap} Pu-240	1.13 ± 2.2 %	1.16 ± 2.2 %	1.07 ± 2.2 %	1.09 ± 2.2 %	0.99 ± 2.2 %	1.02 ± 2.2 %	**12.3 %**
$\sigma_{n,2n}$ Pu-240	1.15 ± 20.0 %		0.49 ± 2.2 %		0.60 ± 20.0%		**13.6 %**
σ_{cap} Pu-241	1.21 ± 4.1 %	-	1.03 ± 4.1 %	-	0.89 ± 4.1 %	-	**14.2 %**
σ_{cap} Pu-242	1.19 ± 3.5 %	-	1.11 ± 3.5 %	-	1.08 ± 3.5 %	-	**13.3 %**
σ_{cap} Np-237	-	0.95 ± 3.6 %	-	0.98 ± 3.6 %	-	0.91 ± 3.6 %	**6.4 %**
σ_{cap} Am-241	1.05 ± 1.7 %	1.07 ± 1.7 %	0.92 ± 1.7 %	0.94 ± 1.7 %	0.87 ± 1.7 %	0.90 ± 1.7 %	**10.6 %**
σ_{cap} Am-243	0.99 ± 5.0 %	-	0.59 ± 5.0 %	-	0.82 ± 5.0 %	-	**18.7 %**

C/E Values of Final Concentrations in the TRAPU Experiment Using JEF-2 Data.

Isotope	TRAPU-1	TRAPU-2	TRAPU-3
U-234	0.97± 3.9 %	1.00± 3.8 %	1.04± 4.6 %
U-235	0.99± 0.4%	1.00± 0.4 %	1.00± 0.4 %
U-236	0.98± 0.8 %	1.00± 1.0 %	1.00± 0.9 %
Np-237	0.70± 6.8 %	0.71± 3.3 %	0.69± 3.2 %
Pu-238	1.02± 1.5 %	1.03± 1.0 %	1.08± 1.6 %
Pu-239	1.02± 0.6 %	1.01± 0.5 %	1.01± 0.4 %
Pu-240	1.02± 0.6 %	1.00± 0.6 %	0.99± 0.6 %
Pu-241	1.09± 0.6 %	1.03± 0.6 %	1.06± 0.6 %
Pu-242	1.15± 0.8 %	1.07± 0.6 %	1.04± 0.6 %
Am-241	0.96± 3.2 %	0.96± 3.9 %	0.96± 2.6 %
Am242M	1.08± 3.8 %	1.11± 4.3 %	1.10± 3.1%
Am-243	1.18± 2.6 %	1.13± 3.1 %	1.15± 2.5 %
Cm-242	1.10± 3.9 %	1.07± 3.1 %	1.07± 2.7 %
Cm-243	-	0.88± 3.1 %	0.89± 3.2 %
Cm-244	1.20± 2.1 %	1.32± 2.3 %	1.33± 1.8 %

C/E Values of Final Concentrations in the TRAPU Experiment Using ENDF/B-VI Data.

Isotope	TRAPU-1	TRAPU-2	TRAPU-3
U-234	0.96± 3.9 %	0.99± 3.8 %	1.03± 4.6 %
U-235	0.99± 0.4%	1.01± 0.4%	1.01± 0.4%
U-236	1.01± 0.8 %	1.03± 1.0 %	1.02± 0.9 %
Np-237	0.75± 6.8 %	0.75± 3.3 %	0.73± 3.2 %
Pu-238	0.96± 1.5 %	0.97± 1.0 %	0.99± 1.6 %
Pu-239	1.03± 0.6 %	1.02± 0.5 %	1.02± 0.4 %
Pu-240	1.02± 0.6 %	1.00± 0.6 %	1.00± 0.6 %
Pu-241	1.07± 0.6 %	1.03± 0.6 %	1.05± 0.6 %
Pu-242	1.08± 0.8 %	1.03± 0.6 %	1.02± 0.6 %
Am-241	0.99± 3.2 %	0.99± 3.9 %	1.00± 2.6 %
Am242M	0.91± 3.8 %	0.94± 3.1 %	0.93± 3.1 %
Am-243	1.05± 2.6 %	1.02± 3.9 %	1.06± 2.5 %
Cm-242	1.02± 3.9 %	1.00± 3.1 %	1.00± 2.7 %
Cm-243	-	0.51± 3.1 %	0.52± 3.2 %
Cm-244	0.66± 2.1 %	0.73± 2.3 %	0.75± 1.8 %

Sensitivity, expressed in percent of variation, of basic cross section to isotope builds up in the TRAPU-I irradiation experiment.

Basic Data	Isotope build-up						
	^{234}U	^{235}U	^{236}U	^{237}Np	^{238}Pu	^{239}Pu	^{240}Pu
$^{234}U\ \sigma_{cap}$	-11.7	0.1					
$^{234}U\ \sigma_{fis}$	-6						
$^{235}U\ \sigma_{cap}$		-10.8	91	12.8	0.7		
$^{235}U\ \sigma_{fis}$	0.4	-37.8	-17.1	-1.6			
$^{236}U\ \sigma_{cap}$			-6	13.6	0.8		
$^{238}U\ \sigma_{cap}$				-2.5	0.2	26.8	3.8
$^{238}U\ \sigma_{(n,2n)}$	0.3			84.4			
$^{237}Np\ \sigma_{cap}$	0.3			-14.5	7.2		
$^{238}Pu\ \sigma_{cap}$	-0.4				-7.3		
$^{238}Pu\ \sigma_{fis}$	-0.8				-13.9		
$^{238}Pu\ \lambda$	9.9				-1.2		
$^{239}Pu\ \sigma_{cap}$						-8.9	25.2
$^{239}Pu\ \sigma_{fis}$					-0.4	-30	-4.2
$^{240}Pu\ \sigma_{cap}$					2.3		-10.2
$^{240}Pu\ \sigma_{fis}$			-0.1				-6.4
$^{241}Pu\ \sigma_{cap}$					-0.4		
$^{241}Pu\ \lambda$	0.7			0.6	19.6		
$^{241}Am\ \sigma_{cap}$	1.5			0.2	35.3		

Sensitivity, expressed in percent of variation, of basic cross section to isotope builds up in the TRAPU-I irradiation experiment.

Basic data	Isotope build-up							
	^{241}Pu	^{242}Pu	^{241}Am	^{242}Am	^{243}Am	^{242}Cm	^{243}Cm	^{244}Cm
^{240}Pu σ_{cap}	47.7	10.6	16.5	6.9	4.8	8.6		2.6
^{241}Pu σ_{cap}	-7.7	36.4	-2.7	-1.1	23	-1.4	-0.8	16.6
^{241}Pu σ_{fis}	-35.6	-7.9	-12.3	-5.1	-3.6	-6.4	-3.9	-2
^{241}Pu λ	-7.8	-0.9	63.8	50.6		54.5	47.5	0.2
^{242}Pu σ_{cap}		-7.6			94.5		-0.2	96
^{241}Am σ_{cap}		1.8	-26.4	82.4	2.2	78.8	85.5	1.9
^{242}Am σ_{fis}				-26.3	-0.2			-0.1
^{243}Am σ_{cap}					-14.8			89.3
^{242}Cm σ_{cap}						-3.2	97.5	-0.3
^{242}Cm λ						-103.4	-52.2	0.2
^{243}Cm σ_{fis}							-20.1	

C/E Values of Final Concentrations in the TRAPU Experiment Using JEF-2 Data from Adjustment With Constraint on Critical Mass.

Isotope	TRAPU-1	TRAPU-2	TRAPU-3
U-234	0.97	0.99	1.03
U-235	0.98	1.00	1.00
U-236	0.99	1.01	1.01
Np-237	0.98	0.97	0.91
Pu-238	1.00	1.00	1.00
Pu-239	1.02	1.00	1.00
Pu-240	1.02	1.00	1.00
Pu-241	1.02	0.99	1.00
Pu-242	1.05	1.02	1.01
Am-241	0.98	0.99	1.00
Am242M	0.98	1.01	1.00
Am-243	1.00	0.98	1.02
Cm-242	0.98	0.96	0.96
Cm-243		0.98	0.98
Cm-244	0.91	1.02	1.04

C/E Values of Final Concentrations in the TRAPU Experiment Using ENDFB-VI Data from Adjustment With Constraint on Critical Mass.

Isotope	TRAPU-1	TRAPU-2	TRAPU-3
U-234	0.96	0.99	1.03
U-235	0.99	1.01	1.01
U-236	0.99	1.01	1.01
Np-237	0.97	0.96	0.90
Pu-238	1.01	1.00	1.03
Pu-239	1.02	1.00	1.00
Pu-240	1.01	1.00	1.00
Pu-241	1.02	0.99	1.01
Pu-242	1.04	1.01	1.01
Am-241	0.97	0.97	0.98
Am242M	0.97	1.00	0.98
Am-243	1.02	1.00	1.04
Cm-242	1.02	1.00	1.00
Cm-243		1.02	1.02
Cm-244	0.89	0.99	1.01

Values of nuclear data before and after adjustment

Basic Data	Starting Values		Adjusted Values	
	JEF2.2	ENDF/B-VI	JEF2.2	ENDF/B-VI
$^{234}U\ \sigma_{cap}$	6.533E-01	6.255E-01	6.514E-01	6.199E-01
$^{235}U\ \sigma_{cap}$	5.691E-01	5.815E-01	5.788E-01	5.755E-01
$^{235}U\ \sigma_{fis}$	1.988E+00	1.940E+00	1.979E+00	1.934E+00
$^{236}U\ \sigma_{cap}$	5.873E-01	5.791E-01	5.998E-01	5.941E-01
$^{238}U\ \sigma_{cap}$	2.933E-01	2.965E-01	2.802E-01	2.787E-01
$^{238}U\ \sigma_{(n,2n)}$	1.310E-03	1.420E-03	1.912E-03	1.926E-03
$^{237}Np\ \sigma_{cap}$	1.665E+00	1.726E+00	1.599E+00	1.693E+00
$^{238}Pu\ \sigma_{fis}$	1.128E+00	1.174E+00	1.147E+00	1.128E+00
$^{238}Pu\ \lambda$	2.510E-10	2.510E-10	2.512E-10	2.513E-10
$^{239}Pu\ \sigma_{cap}$	5.497E-01	5.289E-01	5.272E-01	5.140E-01
$^{239}Pu\ \sigma_{fis}$	1.845E+00	1.845E+00	1.808E+00	1.815E+00
$^{240}Pu\ \sigma_{cap}$	6.200E-01	5.746E-01	5.416E-01	5.261E-01
$^{241}Pu\ \sigma_{cap}$	5.681E-01	4.768E-01	4.496E-01	4.441E-01
$^{241}Pu\ \sigma_{fis}$	2.622E+00	2.550E+00	2.697E+00	2.605E+00
$^{241}Pu\ \lambda$	1.495E-09	1.495E-09	1.523E-09	1.523E-09
$^{241}Am\ \sigma_{cap}$	2.028E+00	1.785E+00	1.805E+00	1.896E+00
$^{242}Cm\ \sigma_{cap}$	5.683E-01	3.473E-01	6.979E-01	6.641E-01
$^{242}Cm\ \lambda$	4.924E-08	4.924E-08	4.915E-08	5.002E-08
$^{242}Pu\ \sigma_{cap}$	5.057E-01	4.461E-01	4.444E-01	4.581E-01
$^{242}Am\ \sigma_{fis}$	3.289E+00	4.118E+00	3.301E+00	3.946E+00
$^{243}Am\ \sigma_{cap}$	1.781E+00	1.116E+00	1.580E+00	1.522E+00

Conclusions and Future Work

- The analysis of PROFIL irradiation experiments using JEF2.2, ENDF/B-V and ENDF/B-VI nuclear data libraries has been performed
- While C/E's obtained for the Uranium isotopes are nearly similar when using both JEF and ENDF/B libraries, the ENDF/B library performs slightly better for the capture cross sections of ^{240}Pu, ^{241}Pu and ^{242}Pu
- Very good results have been obtained using the JEF2.2 data for the ^{241}Am and ^{243}Am radiative captures
- Significant discrepancies on C/E's values have been observed, particularly for the capture cross sections of ^{241}Pu and ^{242}Pu evaluated with JEF2.2 library and the capture cross sections of ^{238}Pu and ^{243}Am evaluated with the ENDF/B libraries
- The uncertainty analysis showed large uncertainty values, particularly for the Plutonium and Americium isotopes

Conclusions and Future Work

- The uncertainties due to data are much larger than the uncertainties due to the experimental accuracy
- The TRAPU C/E's on the final densities of the measured isotopes for the different basic data files have indicated some large discrepancies that a subsequent sensitivity analysis has attributed to specific cross sections of actinides. Among them: ^{238}U (n,2n), capture cross sections of higher plutonium isotopes , ^{241}Am, ^{243}Am, and ^{242}Cm capture cross sections.
- Very useful informations have been gained by performing a simulated adjustment that uses the observed C/E's and the calculated sensitivity coefficients. In particular this adjustment has shown that a really good consistency can be reached on final adjusted cross sections, and associated new C/E's, starting from different data files. The same adjustment has shown that we cannot reach definite conclusions on many fission cross sections (^{238}Pu, ^{241}Pu, ^{242}Am, and ^{243}Cm), because the experiments were not enough sensitive to these parameters.

Conclusions and Future Work

- More investigations need to be done. Calculational details were incomplete for PROFIL-2 and TRAPU because relative to irradiation cycles that were not the actual ones.
- The sensitivity analysis and the simulated adjustment was done at the one group level. In the future a multigroup analysis will provide information by energy range. Also, the adjustment needs to include more critical mass and spectral indices experiment sensitivity coefficients in order to insure a larger consistency among all the data .
- The information from the PROFIL and TRAPU irradiation experiments are relevant to fast energy spectrum reactor types. It would be very useful to add also experiments that provide information in the thermal energy range.